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ROCKWELL INTERNATIONAL ANAHEIM CALIF ELECTRONICS RES--ETC F/6 20/2
GROWTH-INDUCED ANISOTROPY IN YTTRIUM IRON GARNET FILMS GROWN BY--ETC(U)
1975 M T ELLIOTT, H L GLASS F44620-75-C-0045

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1. REPORT NUMBER AFOSR - TR - 77 - 085 L		2. GOVT ACCESSION NO.		3. RECIPIENT'S CATALOG NUMBER	
4. TITLE (and Subtitle) GROWTH-INDUCED ANISOTROPY IN YTTRIUM IRON GARNET FILMS GROWN BY LIQUID PHASE EPITAXY.				5. TYPE OF REPORT & PERIOD COVERED INTERIM rept. g	
7. AUTHOR(s) Elliott, M. T. / Elliott Glass, H. L. / Glass				8. CONTRACT OR GRANT NUMBER(s) F44620-75-C-0045	
9. PERFORMING ORGANIZATION NAME AND ADDRESS Rockwell International Electronics Research Division Anaheim, Calif 92803				10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS 61102F 17 16/2306/81	
11. CONTROLLING OFFICE NAME AND ADDRESS AFOSR/NE Bldg 410 Bolling AFB DC 20332				12. REPORT DATE 1976	
14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office)				13. NUMBER OF PAGES 2	
				15. SECURITY CLASS. (of this report) UNCLASSIFIED	
				15a. DECLASSIFICATION/DOWNGRADING SCHEDULE	
16. DISTRIBUTION STATEMENT (of this Report) Approved for public release; distribution unlimited					
17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)					
18. SUPPLEMENTARY NOTES Proceedings of AIP Conference No. 29 (21st Annl Conf-Philadelphia) Magnetism and Magnetic Materials-1975, American Insti of Physics p.115-116					
19. KEY WORDS (Continue on reverse side if necessary and identify by block number)					
20. ABSTRACT (Continue on reverse side if necessary and identify by block number) Growth-induced anisotropies were measured by ferromagnetic resonance techniques in LPE YIG films which contained appreciable concentrations of impurity Pb ions. Values of K_1 as large as 2.7×10^6 erg/cm ³ were found. The growth-induced anisotropy could be removed by annealing. 19000 GROWTH-INDUCED ANISOTROPY					

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AFOSR - TR - 77 - 035 E

Reprinted from

AIP Conference Proceedings
No. 29

Magnetism and Magnetic Materials-1975

(21st Annual Conference-Philadelphia)

Edited by
J. J. Becker, G. H. Lander, J. J. Rhyne



American Institute of Physics

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GROWTH-INDUCED ANISOTROPY IN YTTRIUM IRON GARNET FILMS GROWN BY LIQUID PHASE EPITAXY*

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ABSTRACT

Growth-induced anisotropies were measured by ferromagnetic resonance techniques in LPE YIG films which contained appreciable concentrations of impurity Pb ions. Values of K_u^G as large as $2.7 \times 10^4 \text{ erg/cm}^3$ were found. The growth-induced anisotropy could be removed by annealing.

INTRODUCTION

Growth-induced magnetic anisotropy is an important and extensively studied property of flux-grown magnetic garnet crystals. It is an especially important property in LPE (liquid phase epitaxy) garnet films which are used in magnetic bubble memory devices. For such applications the growth-induced contribution may be the major source of the uniaxial magnetic anisotropy that is required for the existence of bubble domains.

Bubble garnet compositions generally include Eu or Sm along with one or more smaller rare-earths (or Y). In these materials the large growth-induced anisotropy has been accounted for by models which invoke a partial ordering of the different rare-earth ions on dodecahedral c-sites along with magnetic interactions between the Eu or Sm and neighboring Fe ions¹⁻³. Growth-induced anisotropy has also been observed in mixed garnets and substituted garnets with no magnetic rare-earths present. In these garnets the anisotropy has been attributed to ordered distortions of the crystal field which induce single ion anisotropy in Fe ions. The ordered distortions could arise from ordering of the rare-earth ions in mixed garnets⁴ or of substituent ions in substituted garnets⁵.

Growth-induced anisotropy has also been observed in single rare-earth (or Y) iron garnets. In these materials, the anisotropy has been associated with the incorporation of Pb, as an impurity, from the PbO-based fluxes employed in LPE growth. For Eu-iron garnet ($\text{Eu}_3\text{Fe}_5\text{O}_{12}$) the Pb-induced anisotropy is reported to be similar to the growth-induced anisotropy observed in mixed garnets containing Eu⁶. The same model, partial ordering of Eu on c-sites and magnetic interactions between Eu and Fe, has been invoked to explain the observations. For Y-iron garnet (YIG) there are contradictions in the literature. Some investigators report appreciable growth-induced anisotropy when Pb is incorporated⁷, while other investigators say the effect is very small⁸. In this paper we present some results of our measurements of growth-induced anisotropy in LPE YIG films containing Pb. We will show that the Pb-induced uniaxial anisotropy can be as large as that in mixed garnets containing Eu.

EXPERIMENT AND RESULTS

YIG films a few μm thick were grown on (111) oriented GGG (gadolinium gallium garnet) substrates by the isothermal dipping method of LPE. A $\text{PbO-B}_2\text{O}_3$ flux was used and film growth was carried out while the horizontally held substrates were rotated around a vertical axis. Melt compositions and growth conditions were similar to those described in our recent publication on Pb incorporation in LPE YIG⁸. The incorporation of Pb was controlled by selection of the film growth temperature. The Pb concentration in the films was determined indirectly by X-ray diffraction measurements of the film/substrate lattice misfit from which the film lattice parameter was calculated. The increase of the film lattice parameter (δa_f) relative to the value 12.376\AA for pure YIG

was used as the measure of Pb concentration; the increase being $0.013\text{\AA}/\text{wt \% Pb}$.

Domain patterns in the films were observed using a polarizing microscope. With this technique, stripe domains were visible by the differential Faraday rotation when the films exhibited an appreciable perpendicular component of magnetization. (However, since the Faraday rotation decreases with increasing Pb concentration and changes sign at $\delta a_f \sim 0.02\text{\AA}$ ^{8,9}, the absence of visible domains does not necessarily imply that the perpendicular component is negligible.) For small values of δa_f the misfit stress in the films would be tensile and magnetostriction would produce an easy axis in the [111] direction of the film normal¹⁰; stripe domains would be visible. For films under small compressive stress, $\delta a_f > 0.007\text{\AA}$, the stress-induced anisotropy was in-plane¹⁰ and domains were not visible or were only faintly visible. However, for large compressive stresses, $\delta a_f \geq 0.016\text{\AA}$, intense stripe domain patterns were observed.

Quantitative measurements of the magnetic anisotropy were made by ferromagnetic resonance on small pieces cut from the samples. From the longitudinal and transverse resonance fields at 9.1 GHz the cubic and uniaxial anisotropy fields were determined¹¹. Using the measured value of film/substrate misfit along with elasticity theory and the magnetostriction coefficient of YIG¹⁰, the stress-induced contribution to the uniaxial anisotropy field was calculated and subtracted off to yield the growth-induced contribution. The growth-induced anisotropy K_u^G was obtained from the corresponding anisotropy field ($2K_u^G/M$) using the magnetization M corresponding to the δa_f value of the film⁸. These values of magnetization were confirmed, where possible, from the position of the bottom of the spin-wave band¹². The results are tabulated below:

Film No.	$\delta a_f (\text{\AA})$	$K_u^G (\text{erg/cm}^3)$
1	8.3×10^{-3}	-0.58×10^4
2	15.6	1.33
3	17.4	1.91
4	23.9	2.70

For film 1 K_u^G is negative, indicating an in-plane (non-uniaxial) anisotropy. For this film and for film 2, which has a positive (uniaxial) K_u^G , no stripe domains were observed in the polarizing microscope.

To supplement the above results, other pieces of the same films were annealed in oxygen for 8 hours at 1175°C to determine whether the growth-induced anisotropy would vanish as it does for mixed garnet films. The annealing resulted in a small amount of stress relief in film 1 and drastic stress relief in the other films. In fact, after annealing, films 2-4 were under tensile stress. From the shift in resonance fields brought about by annealing and taking into account the changes in film stress, the growth-induced anisotropies were redetermined¹¹. For film 1, the values of K_u^G before and after annealing agreed to within 3%; however, for the other films the discrepancies ranged from 15-30%.

DISCUSSION

The results clearly demonstrated that LPE YIG films which contain Pb can exhibit a uniaxial growth-induced magnetic anisotropy which is comparable in magnitude to that observed in mixed iron garnets that contain magnetic rare-earths^{1,13}. This growth-induced anisotropy increases with increasing Pb concentration. Using the value for film lattice expansion of $0.013\text{\AA}/\text{wt \% Pb}$, K_u^G has a value of $1 \times 10^4 \text{ erg/cm}^3$ when the Pb concentration is about 1 wt % or 0.04 atoms per

formula unit. If the growth-induced anisotropy is associated with ordering of Pb ions on dodecahedral c-sites, the relatively low concentration of Pb would imply a high degree of site selectivity compared to rare-earths such as Eu. This is not necessarily unreasonable, since Pb is a much larger ion than any rare-earth and since ionic radius mismatch (between Pb and Y in this case) appears to determine site preference¹³.

Although the growth induced anisotropy correlates with Pb concentration, other factors may also be involved. For example, the incorporation of Pb is accompanied by the appearance of Y on octahedral a-sites which would normally be occupied only by Fe^{8,14}. Since Y ions are much larger than Fe, ordering of Y on a-sites could produce large lattice distortions and single-ion anisotropies. Ordering of Y on a-sites could be linked to ordering of Pb on c-sites.

The incorporation of Pb also requires some charge compensation mechanism, since the Pb ions are divalent substituents for trivalent Y. Ferromagnetic resonance measurements have shown that for $\delta a_f \sim 0.007\text{\AA}$ the room temperature linewidth is a minimum¹⁵. This minimum, 0.15 Oe at 9.5 GHz, corresponds to the intrinsic linewidth of pure, perfect YIG. (For film 1, having $\delta a_f = 0.0083\text{\AA}$, the linewidth at 9.1 GHz was 0.14 Oe.) For larger δa_f , the linewidth increases due to charge compensation effects. The dominant charge compensation mechanism appears to be the creation of tetravalent Fe^{15,16}. Another possible mechanism is the presence of oxygen vacancies. The ordering of oxygen vacancies, or other defects, has been suggested as a source of growth induced anisotropy¹⁷. Since the charge compensating defects (tetravalent Fe or oxygen vacancies) are associated with Pb ions and since Y on a-sites may also be associated with Pb, the anisotropy could be due to ordering of some rather complex clusters. Whatever the ordered entity, the absence of magnetic species other than Fe favors those models which are based upon single-ion anisotropy induced by ordered distortions of the crystal field.

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